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ORIGINAL PAPER

Photocatalytic oxidation of benzene by ZnO coated on glass plates under simulated sunlight

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Abstract

The photocatalytic oxidation of benzene by ZnO nanoparticles coated on glass plates was studied under simulated sunlight. ZnO nanoparticles were coated on three glass plates by heat attachment method. To evaluate the photocatalytic removal of benzene, coated plates were irradiated by metal halide lamp in a rectangular reactor in batch mode. The effect of initial pollutant concentration, temperature, relative humidity, irradiation time, concentration of zinc oxide suspension, were assessed. The surface morphology and structure of ZnO nanoparticles and ZnO coated on glass plates were characterized by scanning electron microscopy, X-ray diffraction and field emission scanning electron microscopy. Sampling and analysis of benzene were performed according to NIOSH method. To analyze the concentration of benzene, gas chromatography with flame ionization detector (GC-FID) was used. Results indicated that photocatalytic process by ZnO under irradiation of metal halide lamp could remove benzene at optimum experimental conditions. Coating of glass plates by ZnO suspension, resulted in 57% removal of benzene as concentration of 50 ppm at 45 °C, and relative humidity of 40% after 240 min irradiation of metal halide lamp. Results indicated that photocatalytic oxidation process by ZnO nanoparticles can be used as a proper and environmentally friendly method for removing low concentrations of benzene from polluted air under simulated sunlight.

Keywords Benzene · Air pollution · Photocatalytic oxidation · ZnO · Simulated sunlight

Introduction

Air pollution caused by volatile organic compounds is one of the serious environmental issues (Huang et al. 2017). In urban areas, the VOCs are commonly emitted from different sources including exhaust from motor vehicles, industrial emissions and gasoline service stations (Esplugues et al. 2010). These compounds are the main precursor for the formation of photochemical smog, tropospheric ozone and secondary aerosols (Piumetti et al. 2015).

Among various VOCs, aromatic hydrocarbons especially the benzene series are the most compound of VOCs in polluted urban atmosphere that are highly toxic and harmful to human health. Breathing these compounds can cause adverse impacts on the liver, kidneys, heart, bone marrow, and central nervous system (ATSDR 2007, 2017). The international agency for research on cancer has considered benzene as a carcinogenic compound. Therefore, removing VOCs pollution from the environment is essential to improve the air quality (IARC 2012).

There are various technologies to control VOCs emissions such as physical adsorption, chemical absorption, membrane separation, catalytic combustion, and biological treatment. However, these technologies have disadvantages including the production of secondary pollution, high cost, high-energy consumption, and difficult operation (Khan and Ghoshal 2000; Luengas et al. 2015; Zhang et al. 2017). In addition, the mentioned technologies are mostly applicable for removing high concentrations of the contaminants produced by industries (Luengas et al. 2015). Using these technologies in urban area is not viable for removing low concentrations of contaminants.

Accordingly, numerous studies have been conducted to develop techniques, which are both cost-effective and efficient for removing VOCs in urban areas (Boyjoo et al. 2017; Ren et al. 2017). Photocatalytic oxidation is one of the most

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promising technologies to removing organic contaminants in air using semiconductor materials and presence of light, this process can convert many organic contaminants to less dangerous products such as water (H_2O) and carbon dioxide (CO_2) (Hussain et al. 2011; Zhong and Haghighat 2015).

Since the photocatalytic reactions occur on the surface of the photocatalyst, nanosized photocatalyst materials must be coated on the external surface of supporting media (Tasbihi et al. 2015; Zhong and Haghighat 2015). Currently, numerous studies focused on the application of semiconductor materials on different surfaces such as glass, ceramics, color, and cement materials to remove air pollutants (Boonen et al. 2015; Folli et al. 2015; Pacheco-Torgal and Jalali 2011). External surfaces of building are ideal places to be coated by photocatalyst to remove pollutants in the presence of sunlight (Zhong and Haghighat 2015). Most studies on the photocatalytic processes have been focused on the degradation of VOCs using TiO_2 (Tejasvi et al. 2015; Yao and Yeung 2011). However, it can be activated by ultraviolet (UV) light, which limits the use of sunlight as an energy source for photocatalytic reactions under outdoor environment (Lan et al. 2017). Zinc oxide (ZnO) is a semiconductor material with a large excitation binding energy (60 meV) and similar band gap energy to TiO_2 (3.2 eV), but shows higher performance due to adsorption of large fraction of solar spectrum (Ong et al. 2018). In this study, benzene was chosen as representative aromatic hydrocarbon in air, the photocatalytic efficiency of ZnO nanoparticles coated on glass plates for removal of this pollutant under simulated sunlight irradiation using metal-halide lamp was investigated. The effect of various operating conditions, such as initial benzene concentration, temperature, relative humidity, irradiation time, concentration of ZnO suspension, on degradation of pollutant was assessed.

Materials and methods

Materials

The chemical compounds used in this study included benzene (purity 99.55, Code: B0143) was purchased from Samchun Co. (South Korea), it was used to synthesize the standard solution and prepare artificial polluted air entering the reactor. In addition, to extract benzene from charcoal tube, carbon disulfide (CS_2) prepared by Darkozist industrial and mineral research center was used. The ZnO nanoparticles was prepared from US Research Nanomaterials.

Coating of ZnO on glass plates

To immobilized ZnO powders on the glass plates (22*25, 12*25, 17*25 cm^2), heat attachment method was used (Behnajady et al. 2009). In the first step, ZnO suspension was prepared by adding ZnO powders to distilled water across different concentrations varying 3–12 g/L. In the second step, the prepared suspension was stirred for 30 min on a magnetic stirrer. Then, it was sonicated in an ultrasonic bath (Elmasonic S 80/H, frequency 37 kHz) to completely separate ZnO nanoparticles and obtain a more uniform solution.

Before immobilization, the mentioned glass plates were washed in a NaOH (0.01 M) solution to elevate OH groups and for better contact between ZnO and the plates. They were then washed by distilled water. Afterward, the prepared ZnO suspension was poured on the glass plates. The coated plates were first dried at 120 °C for 1 h in an oven and then calcinated in an electronic furnace (Fanazma Gostar Co., Iran, frequency 50 Hz) for 3 h at 450 °C. The immobilization process was performed three times to increase the loaded ZnO on the plates.

Photocatalytic experiments

The photocatalytic oxidation of benzene was carried out in a rectangular reactor made of Plexiglass with a volume of 15.6 L (30 cm*26 cm*20 cm). To uniform the airflow inside the reactor, a fan was installed in its upper part. The irradiation source was a 150 W metal halide lamp (345–800 nm), which was placed in the central upper part of the reactor and inside a double-walled quartz jacket. The intensity of illumination was measured by a luxmeter as 36,200 lx. To control the temperature of the experiments, the reactor was placed inside an ice bath. Furthermore, to prevent the effect of other lights in the laboratory environment and focus the whole lamp radiation into the reactor, the reactor was covered with aluminum sheet.

To supply the air entering the reactor, an air pump (Resun AC-9906) was used. The pump's airflow was divided into two parts, part of it entered the impinger containing the pollutant liquid to supply contaminated air. The rest of the flow produced by the air pump was entered into the humidifier.

The vapors produced from the impinger and the humidifier entered the mixing chamber (Mariotte's bottle with a volume of 15 L) equipped with a stirrer to uniform the inlet flow to the reactor. The flow resulting from the mixing chamber entered the reactor after recontrolling the flow rate by the rotameter. All of the experiments were performed under a laboratory hood to better control the environmental conditions of the reaction and prevent contamination of the laboratory environment. Figure 1 illustrates the schematic diagram of the experimental setup used in this study.

In all experiments, after the time which had been obtained experimentally considering the flow rate entering the reactor and the volume of reactor, the inlet and outlet of the reactor were closed, and after reaching equilibrium of the flow inside the reactor, the reactor was sampled. Samples were

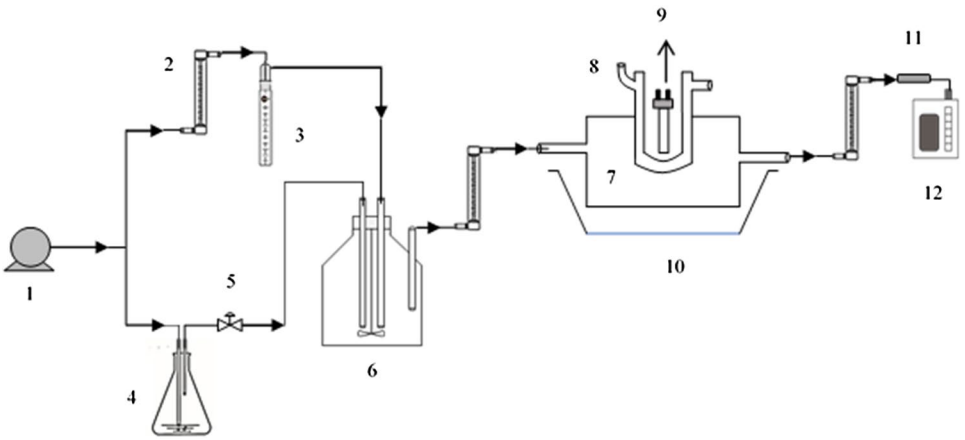

Fig. 1 A schematic diagram of experimental setup. (1) air pump, (2) rotameter, (3) impinger containing pollutant, (4) humidifier, (5) control valve, (6) mixing chamber, (7) photoreactor, (8) double-walled quartz jacket, (9) metal halide lamp, (10) ice bath, (11) charcoal tube, (12) sampling pump

collected with SKC charcoal tube using a personal sampling pump. To analyze the concentration of benzene, gas chromatography with flame ionization detector (GC-FID) (CHROM PACK Co., CP 9001) was used. Sampling and analysis of the flow containing the pollutant were performed according to instruction 1501 NIOSH.

To evaluate the photolysis of the pollutant and adsorption of the pollutant on glass plates, a set of controlling experiments were conducted, (1) in the presence of light and absence of catalytic plates and (2) in the absence of light and presence of catalytic plates. Furthermore, photocatalytic removal of pollutants was evaluated under similar conditions with control experiments conditions.

Catalyst characterization

The structural properties of the surface and mean size of the nanoparticles were determined by scanning electron microscopy (SEM). The crystalline nature and purity of ZnO were specified by x-ray diffraction (XRD). Furthermore, after coating the nanoparticles on the glass plates, the structural properties of the immobilized nanoparticles were characterized using field emission scanning electron microscopy (FE-SEM) (MIRA3 TESCAN).

Results and discussion

Characterization

The XRD pattern of ZnO powders is shown in Fig. 2. Nine diffraction peaks of ZnO nanoparticles are found at 37.132°–86.896° corresponding to ZnO crystal phase (JCPDS 5-0664). No other peaks have been determined from other phases, indicating that the crystalline phase is individually related to zinc oxide. The result of SEM image (Fig. 3) shows that the size of nanoparticles is around 20 nm. Therefore, the surface–volume ratio of nanoparticles has been high, and the specific surface area of the nanoparticles was 20–60 m^2/g. The results of FE-SEM analysis (Fig. 3) which has been performed on the nanoparticles coated on the glass surface indicate suitable distribution of nanoparticles on the glass plate.

Comparison of photocatalytic oxidation with controlling experiments

The results of Fig. 4 represent that irradiation of metal halide lamp only removed 16% of benzene. In addition,

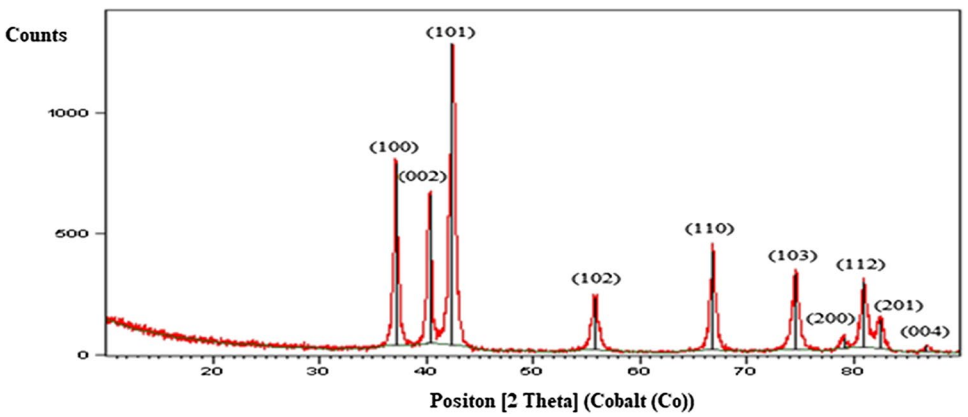

Fig. 2 XRD pattern of ZnO

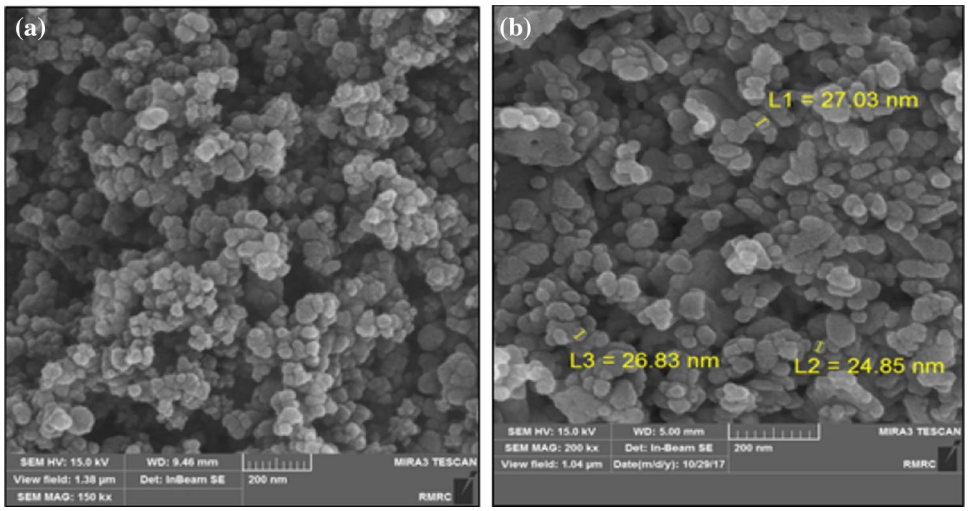

Fig. 3 **a** SEM image of ZnO nanoparticles, **b** FE-SEM image of ZnO coated on glass plate

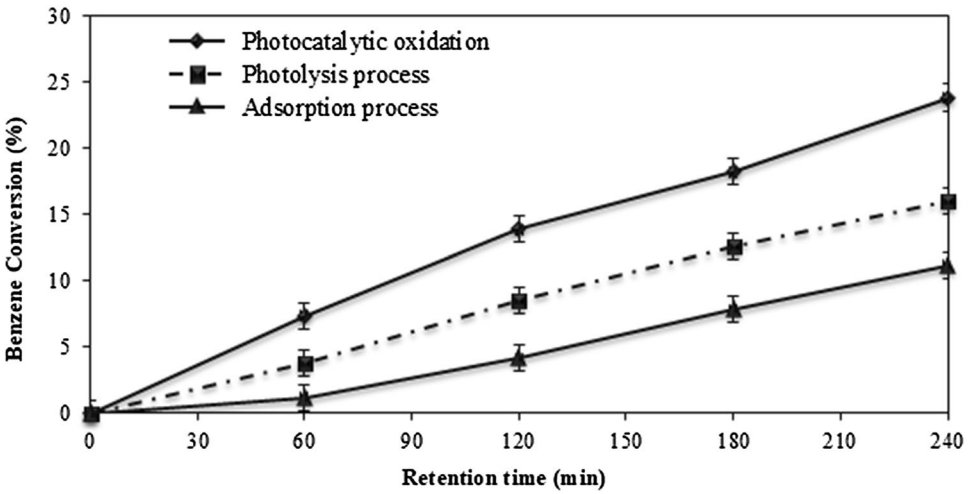

Fig. 4 Benzene conversion by photocatalytic oxidation, photolysis, and adsorption process. Initial benzene concentration 50 ppm, at 25 °C, relative humidity 10%, and ZnO suspension dosage 10 g/L

using catalytic plates alone under dark condition led to 11.15% of benzene adsorbed. However, photocatalytic oxidation under the same condition illustrated that 23.80% of benzene removed after 240 min. These results indicated that under irradiation of metal halide lamp, the presence of ZnO could accelerate the reaction and more pollutants removed. Photocatalytic oxidation is a superficial process that occurred in five steps. When the pollutant is in contact with the photocatalyst, it is absorbed on the surface of active sites of the catalyst. This process is described in the following equations:

$$ZnO + H_2O \longrightarrow ZnO\_H_2O \quad (1)$$

$$ZnO + O_2 \longrightarrow ZnO\_O_2 \quad (2)$$

$$ZnO + C_6H_6 \longrightarrow ZnO\_C_6H_6 \quad (3)$$

The photocatalytic process occurred at the photocatalyst surface with the reaction of pollutants with hydroxyl radicals in the presence of a light. This process involves two steps, first the electron–hole pair is created and then the oxidation of the contaminants occurs (Ong et al. 2018).

$$ZnO \xrightarrow{h\nu} ZnO\,(e^- + h^+_{vb}) \quad (4)$$

$$ZnO\,(e^- + h^+_{vb}) \longrightarrow ZnO + \text{heat} \quad (5)$$

$$ZnO\,(h^+_{vb}) + H_2O \longrightarrow ZnO + H^+ + OH^- \quad (6)$$

$$ZnO + OH^- \longrightarrow ZnO + OH^{\bullet} \quad (7)$$

$$ZnO\left(e^-_{cb}\right) + O_2 \longrightarrow ZnO + O_2^{\bullet -} \quad (8)$$

$$O_2^{\bullet -} + H^+ \longrightarrow HO_2^{\bullet} \quad (9)$$

$$HO_2^{\bullet} + HO_2^{\bullet} \longrightarrow H_2O_2 + O_2 \quad (10)$$

$$ZnO\left(e_{cb}^{-}\right) + H_2O_2 \longrightarrow OH^{\cdot} + OH^{-} \quad (11)$$

$$H_2O_2 + O_2^{\bullet -} \longrightarrow OH^{\cdot} + OH^{-} + O_2 \quad (12)$$

$$H_2O_2 + h\nu \longrightarrow 2\,OH^{\bullet} \quad (13)$$

$$\text{Organic pollutant} + OH^{\bullet} \longrightarrow \text{Intermediates} \quad (14)$$

$$\text{Intermediates} \longrightarrow CO_2 + H_2O \quad (15)$$

Huiling Huang et al. (2015) observed that the photocatalytic removal of benzene by TiO_2 under VUV irradiation was 50%, but irradiation of VUV alone resulted in 27% removal of this pollutant (Huang et al. 2015). Masoud Rismanchian et al. (2014) showed that irradiation of UV lamp alone and using uncoated nickel foam resulted in 8% toluene removal, but photocatalytic oxidation of toluene led to 50% of toluene removed after 270 min (Rismanchian et al. 2014).

Effect of initial concentration of the pollutant

The effect of initial concentration on photocatalytic oxidation of benzene is shown in Fig. 5. The results show that with increasing of the initial pollutant concentration, the photocatalytic oxidation efficiency decreased. In this study, increasing of concentration from 50 to 240 ppm, reduction of efficiency was observed for removal of benzene from 17 to 2.94%.

The effect of the inlet concentration of pollutant in the field of photocatalytic oxidation has been studied by different researchers (Ma et al. 2015; Rezaee et al. 2014). Most studies have concluded that increasing the pollutant concentration results in decreased removal efficiency of the pollutant (Chen et al. 2018; Nath et al. 2014). Abbas Rezaee et al. (2008)indicated that the elevation of toluene concentration caused reduced photocatalytic efficiency by TiO_2 nanoparticles (Rezaee et al. 2008). Chen et al. (2018) showed that benzene removal efficiency by BiVO4/α-Fe_2O_3 composite decreased from 66.87 to 33.82% when increasing the concentration from 100 to 600 mg/m^3 (Chen et al. 2018).

The effect of higher concentration of pollutants on photocatalytic oxidation can be attributed to the decrease of active sites on photocatalytic surface (Lyu et al. 2014; Yu and Brouwers 2009). As the pollutants concentration increase, more pollutant molecules could adsorb on the photocatalyst surface. Therefore, the need to active oxidant species such as hydroxyl radical to degrade this high concentration of pollutants also increase (Mo et al. 2009). In addition, the excess pollutant molecules on the photocatalyst surface prevent adsorption of optical photons by the catalyst. Thereby, the active oxidant species such as hydroxyl radicals (OH·) and superoxide anions ($O_2^{\bullet -}$) are inadequate for photocatalytic oxidation of high concentration of pollutants when the irradiation is constant (Mamaghani et al. 2017).

Effect of temperature

In the present study, experiments were conducted across different temperatures including 19, 25, 35, and 45 °C. The results of Fig. 6 show that increasing the temperature has led to enhanced efficiency of the photocatalytic oxidation in the removal of benzene, the increase of temperature to 45 °C resulted in 31.13% removal for benzene.

Generally, surface reaction includes three main steps: adsorption of the pollutant on the catalyst surface, chemical reaction, and desorption of the product off the catalyst surface (Obee and Hay 1997). Temperature affects the

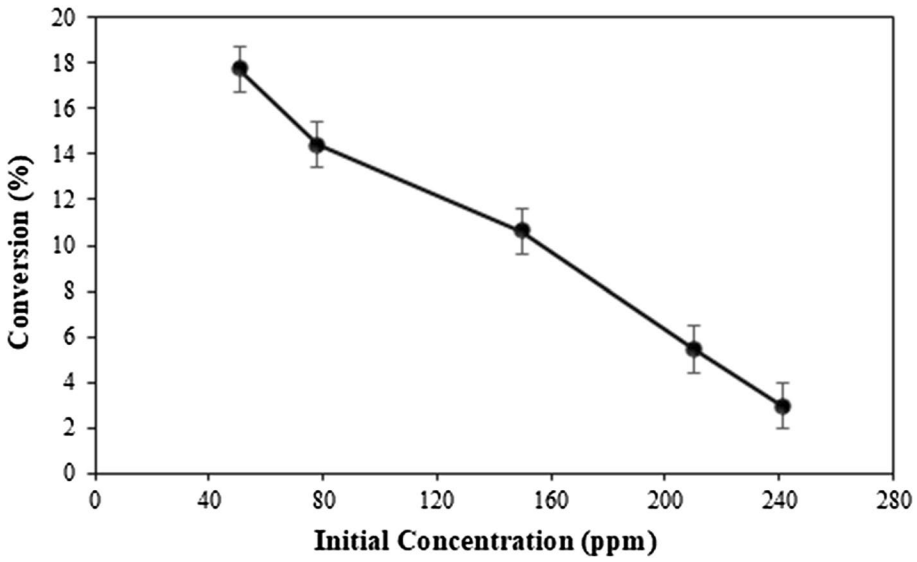

Fig. 5 Effect of initial pollutant concentration at 25 °C, relative humidity 10%, irradiation time 120 min, and ZnO suspension dosage 10 g/L

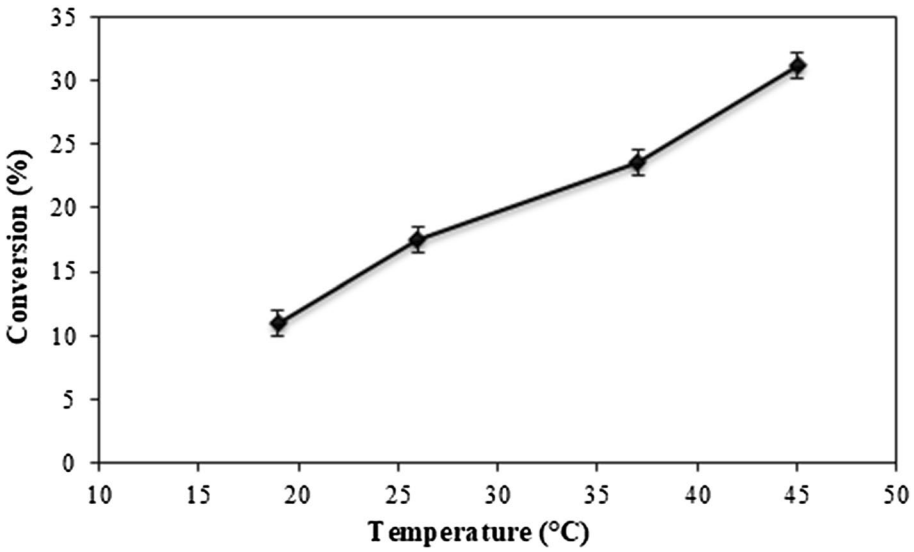

Fig. 6 Effect of temperature. Initial benzene concentration 50 ppm, relative humidity 10%, irradiation time 120 min, and ZnO suspension dosage 10 g/L

photocatalytic oxidation reaction and adsorption of pollutant on photocatalyst surface (Mo et al. 2009). Increasing the temperature, the internal energy of benzene molecules increases and their irregular motion is intensified. According to Arrhenius temperature dependence coefficient, k, the increase in temperature has a positive impact on photocatalytic oxidation kinetic reaction (Yu and Kim 2013).

$$k = f\left(\exp\left(\frac{-E}{RT}\right)\right) \tag{16}$$

where E is an apparent activation energy, normally greater than 0; T is the temperature and R the gas constant (8.314×10^{-3} kJ. mol^{-1}. K^{-1}).

At high temperatures, the rate of chemical reaction and desorption of the product increases. However, temperature elevation also causes reduction of rate of adsorption of the pollutant on the catalyst surface (Mo et al. 2009). The adsorption equilibrium coefficient, K, was calculated according to Eq. 2 (Yu and Kim 2013).

$$K = f\left(\exp\left(\frac{-H}{RT}\right)\right)/\sqrt{T} \tag{17}$$

where H is the change in enthalpy adsorption for the adsorbed pollutant. In the present study, as typical air temperatures were investigated, the removal efficiency of the pollutant increased with increasing the temperature. It can be stated that reduced adsorption of the reactant at the optimal temperature had no effect on the general efficiency of the photocatalytic process.

Wei Cai et al. (2014) studied the removal of acetone and ethanol by TiO_2 across different temperatures including 14, 31, and 43 °C. They observed that higher temperatures led to enhanced acetone and ethanol removal (Cai et al. 2014). In the research by Fumihide Shiraishi et al. (2005) to evaluate the efficiency of removing formaldehyde by TiO_2 across the temperatures of 45, 55, 70, and 90 °C, it was observed that the temperature 45 °C had maximum efficiency in formaldehyde removal. As mentioned previously, the reason was less adsorption of the pollutant at higher temperatures on the catalyst surface (Shiraishi et al. 2005). Wu et al. (2005) studied the effect of temperature from 100 to 200 °C on photocatalytic oxidation of benzene by TiO_2. Results showed that the removal of benzene increased as the temperature increased from 100 to 160 °C, then decreased to 240 °C (Wu et al. 2005).

Effect of relative humidity

The effect of relative humidity on photocatalytic oxidation of benzene was examined using different RH including 10, 30, 40, and 60%. The results in Fig. 7 indicate that with increasing humidity, the removal efficiency of the pollutant has increased. However, elevation of humidity from 40 to 60% has led to diminished photocatalytic efficiency.

Since the surface of photocatalyst under the irradiation of light is hydrophilic, water molecules adsorb on the surface of photocatalyst and would have positive and negative effect on photocatalytic oxidation of pollutants (Huang et al. 2015). As water vapor is a very important source of hydroxyl radicals in photocatalytic oxidation, Presence of water vapor is essential for strengthening the photocatalytic process (Mamaghani et al. 2017).

Nevertheless, excessive water molecules on the catalyst surface form multi-layers of water on the catalyst surface (Mamaghani et al. 2017). Therefore, water molecules inhibit adsorption of the pollutant on the catalyst surface and the pollutants leave the reactor untreated. Mohamad Sleiman et al. (2009) observed that the maximum conversion of photocatalytic removal of toluene using TiO_2 achieved at the humidity of 30%. However, at 50 and 70%, toluene removal

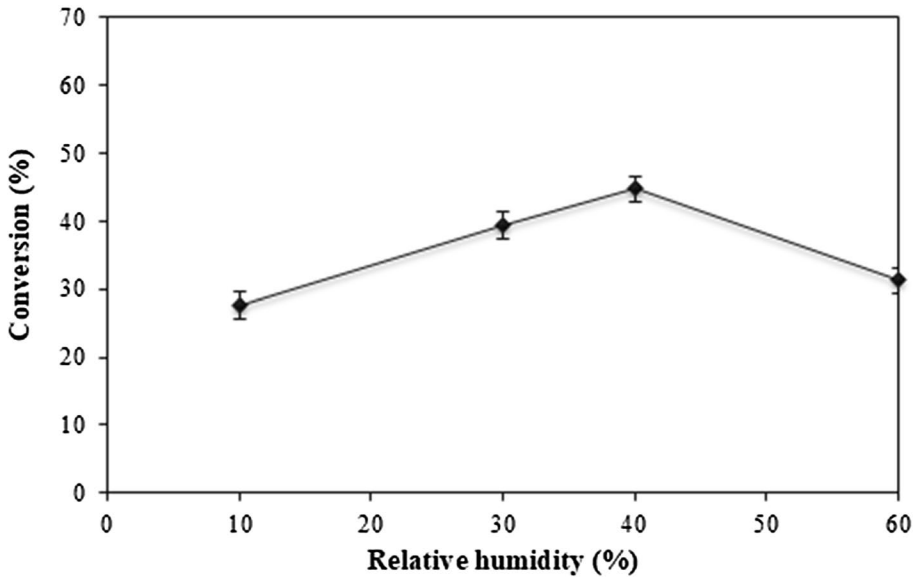

Fig. 7 Effect of relative humidity. Initial benzene concentration 50 ppm, 45 °C, irradiation time 120 min, and ZnO suspension dosage 10 g/L

efficiency decreased (Sleiman et al. 2009). Masoud Rismanchian et al. (2014) studied toluene removal efficiency by TiO_2 nanoparticles coated on nickel metal foam at different humidity 10, 30, 50 and, 80%. They found that elevation of humidity first increased removal efficiency of the pollutant then increasing humidity from 30 to 80% resulted in decreased photocatalytic efficiency (Rismanchian et al. 2014).

Effect of irradiation time

Investigating the effect of irradiation time on the photocatalytic efficiency of ZnO in the removal of benzene indicated that with increasing the irradiation time, the photocatalytic efficiency of ZnO in removal of pollutant has increased. As shown in Fig. 8, at the irradiation time 30 min the removal efficiency of benzene was 2.95%. However, increasing the irradiation to 240 min has increased removal efficiency to 55.23%.

Increasing the irradiation time led to increased photons and thus further hydroxyl radical production. In addition, increased irradiation time to photocatalytic surfaces results in prolonged contact time between the pollutant and catalyst surface. In this way, higher contact times enhance the mass transfer coefficient between the air and photocatalyst surface.

Therefore, more molecules of the pollutant can adsorb on the active sites of the photocatalyst surface and react with the hydroxyl radicals (Lyu et al. 2014).

Studies also suggest enhanced removal efficiency with increasing irradiation time. Masoud Rismanchian et al. (2014) indicated that retention times lower than 100 min did not have a good efficiency in removing toluene by TiO_2

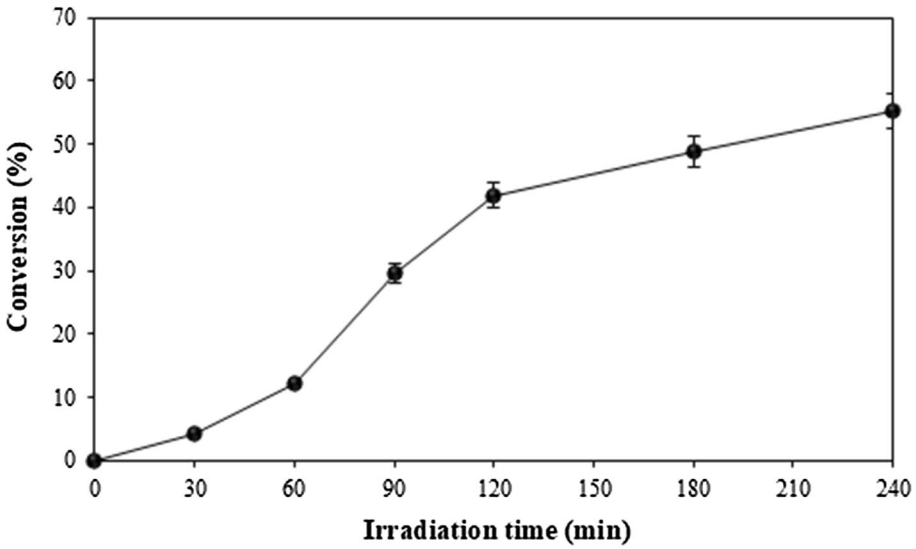

Fig. 8 Effect of irradiation time. Initial benzene concentration 50 ppm, 45 °C, relative humidity 40%, and ZnO suspension dosage 10 g/L

nanoparticles stabilized on nickel metal foam. Nevertheless, with increasing irradiation time to 270 min, almost 55% of toluene was removed (Rismanchian et al. 2014). Gholamhossein Pourtaghi et al. (2009) investigated the photocatalytic oxidation of toluene by TiO_2 at times of 10, 30, and 60 min and at inlet pollutant concentrations of 150, 250, 350, and 450 ppm. They observed that toluene removal efficiency was maximum at the retention time of 60 min and pollutant concentration of 150 ppm (Pourtaghi 2009). Zhi Liu et al. (2012) also showed that retention time affects mineralization of benzene through photocatalytic oxidation process by CdS–TiO_2 coated on fiberglass. Increasing retention time from 30 to 300 min caused enhanced carbon dioxide production resulting from the process (Liu et al. 2012).

Effect of ZnO suspension dosage

Investigating the effect of different concentration of ZnO suspension on the removal of benzene illustrate (Fig. 9) that elevation of ZnO suspension concentration to 12 g/L resulted in 57.68% removal for benzene.

Increasing the ZnO suspension concentration caused increased loading of nanoparticles on the glass surface. Therefore, the surface area of nanoparticles and the number of active sites on the catalyst surface increase which elevates the amount of adsorbed pollutants (Mamaghani et al. 2017).

Behnajady et al. (2009) also demonstrated that increasing the ZnO suspension concentration led to further loading of ZnO on the glass plates, thereby enhancing the pollutant removal efficiency. The results indicated that an increase in the ZnO suspension dosage from 2 to 12 g/L showed that the suspension with the concentration of 12 g/L had the maximum efficiency, though no significant difference was observed between the results obtained from the photocatalytic oxidation with suspension concentrations of 10 and 12 g/L(Behnajady et al. 2009). Morteza Jafarikojour et al. (2015) observed that as the amount of TiO_2 coated on stainless steel sheets increased from 0.274 to 0.925 g/m^2, the degradation of toluene enhanced from 26 to 43% (Jafarikojour et al. 2015). Hisahiro Einaga et al. (2004) also observed similar behavior for photocatalytic oxidation of benzene. Results illustrated that as the amount of TiO_2 loading was changed from 0 to 0.24 g, the averaged catalyst weight was increased to 2 mg/cm^2 and benzene conversion was enhanced (Einaga et al. 2004).

Conclusion

The present study evaluated the efficiency of photocatalytic conversion of benzene by ZnO nanoparticles coated on glass surface under simulated sunlight. Results indicated that with variation of operating conditions including initial pollutant concentration, temperature, relative humidity, irradiation time, concentration of ZnO suspension, photocatalytic conversion of benzene altered. Generally, it can be stated that the mentioned photocatalytic process had a relatively good efficiency under the controlled experimental conditions. Coating of glass surfaces by ZnO suspension, resulted in 57% removal of benzene as concentration of 50 ppm at 45 °C, and relative humidity of 40% after 240 min irradiation of metal halide light.

However, as under realistic conditions, various parameters affect the process which cannot be considered in the

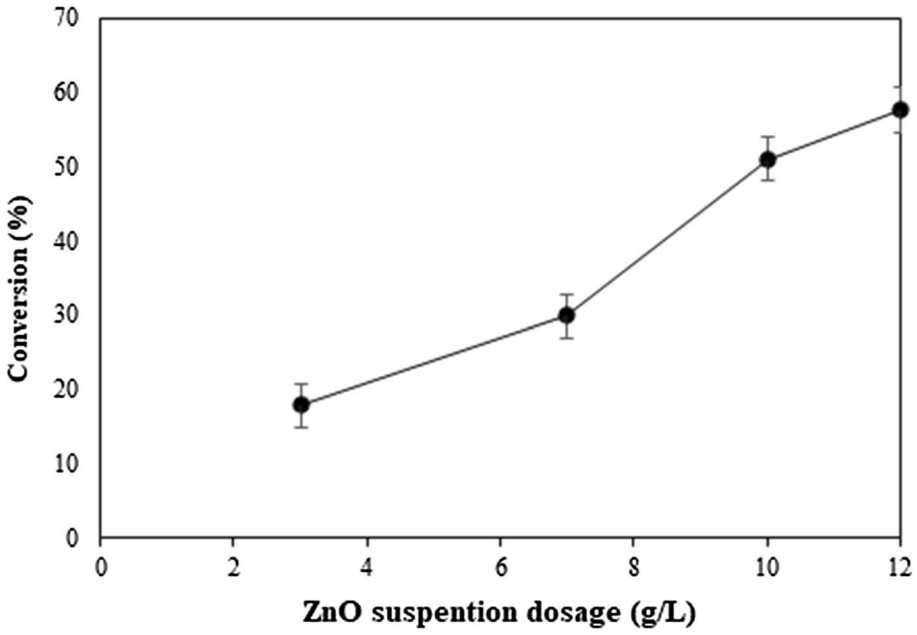

Fig. 9 Effect of ZnO suspension dosage. Initial benzene concentration 50 ppm, 45 °C, relative humidity 40%, and irradiation time of 240 min

experimental medium, the photocatalytic process efficiency may be different under real conditions, suggesting that application of this method requires further investigation. Furthermore, air pollution contains a mixture of contaminant that may inhibit photocatalytic removal of benzene. Therefore, further studies should be conducted regarding removal in a mixture of air pollutants by the photocatalytic oxidation process.

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